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(54) [Title of Invention]

**PROPYLENE POLYMER WHICH IS ACQUIRED
MAKING USE OF POLYMERIZATION METHOD OF
PROPYLENE AND THAT**

(51) [International Patent Classification, 7th Edition]

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[Examiner]

***Sawa **

(57)[Claim(s)]

[Claim 1]

Other than propylene , the;al to ethylene or carbon number 4
or more 12 0.01 - 30 weight % it is possible to include -olefin
as comonomer , with polymerization method of the
propylenic monomer , critical temperature of monomer
component where polymerization temperature exists inside
polymerization system compared to with melting point or
higher of polymer which and is polymerizedhighly, it is
higher than critical pressure of monomer component where
polymerization pressure exists inside aforementioned
polymerization system , polymerization temperature 170 deg
C~250 deg C, polymerization pressure is inside range of 50 -
300 kg/cm ², propylene ittatches on solventless
polymerization under condition and makes feature, the

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polymerization method . of aforementioned propylene

[Claim 2]

Q of polymer is 6 or more , polymerization method . of propylene which is stated in Claim 1

[Claim 3]

polymerization method . of propylene which uses Ziegler catalyst , states in Claim 1

[Claim 4]

polymerization method . of propylene which uses magnesium-bearing catalyst , states in Claim 1

[Claim 5]

polymerization method . of propylene which uses catalyst which prepolymerization is done with vinyl group-containing compound , states in Claim 1

[Claim 6]

melting point of polymer which prepolymerization is done is 200 deg C or greater , polymerization method . of propylene which is stated in Claim 5

[Claim 7]

With polymer which was polymerized with method which is stated in Claim 1, MFR of said polymer is 5 - 1000 g/10 min , it makes feature, propylene polymer .

[Claim 8]

With polymer which was polymerized with method which is stated in Claim 1, Q of said polymer is 6 or more , it makes feature, propylene polymer .

[Description of the Invention]

[0001]

[Technological Field of Invention]

this invention is something regarding polymerization method of propylene .

Furthermore details regard polymerization method of propylene which possesses the high stereoregularity with quite high activity .

[0002]

[Prior Art]

Until recently as method which polymerizes propylene , uses the hexane , heptane etc as solvent gas phase method etc which designates bulk method , inert gas which designates slurry method , propylene itself which as solvent as media is known.

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With these method , it has reached point where polymer where the stereoregularity is high with high activity with performance improvement of catalyst which is used and improvement of polymerization technology , is acquired.

But, concentration of monomer in reaction system in solvent regarding the slurry method is low, in addition, in order because polymerization temperature is low, the polymerization activity is low, to enlarge this there is a limit .

In addition, solvent remaining in polymer , level of smell of polymer there was a problem which deteriorates.

[0003]

On one hand, also bulk polymerization method which polymerizes in liquid state monomer is done, but as for bulk polymerization because polymerization temperature is low, polymerization activity not to be a satisfactory , in addition, energy which is large to separation and reuse of monomer after polymerizing is needed, there was a problem where cost becomes high.

In addition, although there is not a problem etc of above-mentioned solvent regarding gas phase method , problem in regard to or other driving which massive polymer is easy to form is held.

As these problem are solved, furthermore polymerization method which actualizes the high productivity with high activity was desired.

[0004]

[Problems to be Solved by the Invention]

this invention smells with solvent , there is not a formation or other problem of the massive polymer in gas phase method , polymerization activity quite is high, is superior in productivity , it is to offer method which produces polymer where the stereoregularity is high.

[0005]

[Means to Solve the Problems]

You discovered method which can offer polymer which possesses high steric rule with high activity where as for this inventor etc, in order to solve aforementioned problem , as for result of examining the various polymerization method , polymerization stability is high by polymerizing propylene with specific condition , in addition is not so far informed, arrived in this invention .

Namely, it is something where this invention, with polymerization method of propylenic monomer , critical temperature of monomer component where polymerization temperature exists inside polymerization system compared to with melting point or higher of polymer which and is polymerized highly, propylene attaches on solventless

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polymerization under condition which is higher than critical pressure of monomer component where polymerization pressure exists inside the aforementioned polymerization system makes feature, offers polymerization method of the aforementioned propylene.

[0006]

[Embodiment of the Invention]

this invention with temperature which is higher than critical temperature of monomer component which exists inside polymerization system and is polymerized with temperature of the melting point or higher of polymer which is polymerized.

Namely, it makes temperature range of melting point or higher of polymer where polymerization temperature when from critical temperature of propylene which is offered to the polymerization, in addition producing copolymer, is high in comparison with all critical temperature of propylene and other comonomer component (You detail on description below, but the;al of ethylene and carbon number 4 or more - olefin etc) which are used, at same time forms.

[0007]

In addition, polymerization pressure makes high pressure in comparison with the critical pressure of monomer component which exists inside polymerization system.

Furthermore in this invention, "melting point of polymer" with, polymerizing under same condition as condition which is done beforehand actually, as a result it means that melting point of produced polymer which is acquired was measured due to the differential thermal analysis (DSC).

[0008]

When polymerization temperature, it is lower than critical temperature of monomer component which exists inside polymerization system, monomer for polymerization doing, liquefaction because density of monomer becomes high, as benefit of this invention becomes impossible is enjoyed, cost of separation and reuse of monomer becomes high.

Furthermore, when being lower than melting point of polymer, molecular weight distribution which is made objective of this invention it is wide, preferably Q stops being acquired, propylene polymer of 6 or more is not desirable.

[0009]

In addition, when polymerization pressure, being lower than critical pressure of monomer component which exists inside polymerization system, supercritical phenomenon cannot reveal and cannot acquire high activity.

As for polymerization temperature and polymerization pressure homopolymerization or copolymerization?, or

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 45.4kg/cm² □□□□□□45.4kg/cm²
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 deg C~250 deg C□□□□□□50~300kg/cm²
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it changes with molecular weight regulator for example hydrogen or other presence or absence or other condition, but when homopolymer of for example propylene is produced, as for critical temperature of propylene as for melting point of polymer which in 91.4 deg C, is acquired, it differs depending upon level of the stereoregularity of polymer which is acquired, but because they are 160 - 165 deg C generally, It is necessary to polymerize with 165 deg C or greater.

In addition, because as for polymerization pressure, as for critical pressure of propylene they are 45.4 kg/cm² ², it is necessary to polymerize with pressure which is higher than 45.4 kg/cm² ².

[0010]

In addition, regarding to this invention, polymerization temperature 170 deg C~250 deg C, polymerization pressure solventless polymerization is done in inside range of 50 - 300 kg/cm² ².

Regarding to this invention, solventless polymerization, system for slurry polymerization, solution polymerization which uses solvent is something which polymerizes in composition which designates monomer as main component unlike system which advances the polymerization with state which it melts or disperses catalyst and polymer, or including solvent, in finger naming and the polymerization system.

To typical, polymerization advances in monomer composition of supercritical state.

But, it is not something which removes coexistence of solvent of trace, fact that rather it is used for case of catalyst component, additive, monomer or other supply as diluent, or carrier is desirable embodiment.

[0011]

If it is something which it can withstand temperature, pressure or other condition which is adopted for method of this invention as polymerization equipment which can be used with the this invention, there is not especially restriction, be able to use public knowledge polymerization equipment, reaction system it can use loop type reactor etc which circulates with stirring tank, blower which possesses for example stirrer blade-equipped power unit.

[0012]

In addition, regarding to this invention, as monomer which is offered to polymerization, other than propylene, ethylene or carbon number 4 or more, usually the; al to carbon number 12 you can list -olefin as comonomer, can list ethylene, butene -1, 4- methyl -pentene -1, hexene -1, octene -1, decene -1, 3 - methyl -butene -1, etc concretely.

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□□□□□□□□□MFR□5~1000g/10□□□□
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□□□□MFR□ASTMD1238□□□□□□□□
□□□□□□□□(230 deg C□2.16kg□□)□

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□□Q□□Mw/Mn(Mw:□□□□□□□□□□
□□□□□□□□(GPC)□□□□□□□□□□
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Quantity of comonomer is not something which especially is restricted. They are 15 weight % extent from 0.01 - 30 weight %, preferably 0.1 .

[0013]

In addition, following to method of this invention , as for propylene polymer which was polymerized, it is desirable for MFR to be 5 - 1000 g/10 min

Furthermore, this MFR conforming to ASTM D1238, is something which is measured, (230 deg C, 2.16kg load).

[0014]

Furthermore, following to method of this invention , as for propylene polymer which was polymerized, it is desirable for Q to be 6 or more .

When Q is this range, moldability of mechanical strength corresponding and external appearance of molded article are superior, when it is too low, there is a tendency where external appearance of moldability and molded article is inferior.

Furthermore Q is something which is calculated with Mw/Mn (weight average molecular weight , Mn which was measured by Mw :gel permeation chromatograph (GPC) is number average molecular weight which was measured by GPC .).

[0015]

As for catalyst which is used with this invention , it can recognize the effect of this invention , if you obtain with those of option , but because Ziegler catalyst , especially being a catalyst of magnesium bearing type is superior, activity in high temperature , it is desirable.

Concretely, below-mentioned component (A) and component (B), or component (A), component (B) and it is something which consists of combination of component (C).

It is not something where here, "Of combination it consists " with as for notion that where you say, that component which is used is only raising/holding Shimesu thing (namely, component (A) and (B), component (A), (B) and (C)) you say and mean , coexistence of goal-consistent other component is not removed.

[0016]

<component A>component (A) is solid component for Ziegler catalyst which contains titanium , magnesium and halogen as essential ingredient .

Here "It contains as essential ingredient " with may include goal-consistent other element outside raising/holding Shimesu three components to say, these element each one may exist as

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 127405□□□55-147507□□□55-
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 155206□□□57-3803□□□57-
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 121003□□□58-5309□□□58-5310□□□58-
 5311□□□58-8706□□□58-27732□□□58-
 32604□□□58-32605□□□58-
 67703□□□58-117206□□□58-
 127708□□□58-183708□□□58-
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 □Ti(OR□)_{4-q} X_q
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TiCl₄ □

TiBr₄ □

compound of the goal-consistent option , and these element may exist as is connected mutually, it is something which is shown.

solid component itself which includes titanium , magnesium and halogen is something of public knowledge .

for example Japan Unexamined Patent Publication Showa 53-45688 number, same 54 - 3894, same 54 - 31,092, same 54 - 39483, same 54 - 94591, same 54 - 118484, same 54 - 131,589, same 55 - 75411, same 55 - 90510, same 55 - 90511, same 55 - 127405, same 55 - 147507, same 55 - 155003, same 56 - 18609, same 56 - 70005, same 56 - 72001, same 56 - 86905, same 56 - 90807, same 56 - 155206, same 57 - 3803, same 57 - 34103, same 57 - 92007, same 57 - 121003, same 58 - 5309, same 58 - 5,310, same 58 - 5,311, same 58 - 8706, same 58 - 27732, same 58 - 32604, same 58 - 32605, same 58 - 67703, same 58 - 117206, same 58 - 127708, same 58 - 183708, same 58 - 183709, those which are stated in same 59 - 149905, same 59 - 149906 each disclosure etc are used.

[0017]

Regarding to this invention, you can list carbonate etc of magnesium halide , di alkoxy magnesium , alkoxy magnesium halide , magnesium oxyhalide , dialkyl magnesium , magnesium oxide , magnesium hydroxide , magnesium as the magnesium compound which becomes magnesium source which is used.

Desirable ones are magnesium halide , di alkoxy magnesium , alkoxy magnesium halide among these.

[0018]

In addition, as for titanium compound which becomes titanium source , it can increase compound which is displayed with General Formula Ti (OR_{4-q})_q (As for R with hydrocarbon residue , with those of preferably carbon number 1~10 extent , as for the X halogen is shown here, q 0 ≤ shows quantity of q*4.).

As embodiment ,

TiCl₄ ,

TiBr₄ ,

$$\text{Ti}(\text{OC}_2\text{H}_5)_3 \quad \square$$
$$\text{Ti}(\text{OC}_2\text{H}_5)_2\text{Cl}_2 \quad \square$$

$\text{Ti}(\text{OC}_2\text{H}_5)_3$ $\text{Cl}\square\text{Ti}(\text{O}-\text{iC}_3\text{H}_7)\text{Cl}_3$ $\square\text{Ti}(\text{O}-\text{nC}_4\text{H}_9)\text{Cl}_3$ $\square\text{Ti}(\text{O}-\text{nC}_4\text{H}_9)_2\text{Cl}_2$ $\square\text{Ti}(\text{OC}_2\text{H}_5)\text{Br}_3$
 $\square\text{Ti}(\text{OC}_2\text{H}_5)(\text{OC}_4\text{H}_9)_2\text{Cl}$ $\square\text{Ti}(\text{O}-\text{nC}_4\text{H}_9)_3\text{Cl}$ $\square\text{Ti}(\text{O}-\text{C}_6\text{H}_5)\text{Cl}_3$ $\square\text{Ti}(\text{O}-\text{iC}_4\text{H}_9)_2\text{Cl}_2$
 $\square\text{Ti}(\text{OC}_5\text{H}_{11})\text{Cl}_3$ $\square\text{Ti}(\text{OC}_6\text{H}_{13})\text{Cl}_3$ $\square\text{Ti}(\text{OC}_2\text{H}_5)_4$ $\square\text{Ti}(\text{O}-\text{nC}_3\text{H}_7)_4$ $\square\text{Ti}(\text{O}-\text{nC}_4\text{H}_9)_4$ $\square\text{Ti}(\text{O}-\text{iC}_4\text{H}_9)_4$
 $\square\text{Ti}(\text{O}-\text{nC}_6\text{H}_{13})_4$ $\square\text{Ti}(\text{O}-\text{nC}_8\text{H}_{17})_4$ $\square\text{Ti}\square\text{OCH}_2\text{CH}(\text{C}_2\text{H}_5)\text{C}_4\text{H}_9$ \square_4
 $\square\square\square\square\square\square\square\square$

□0019□

$$\begin{array}{l} \square\square\square\text{TiX}\square_4 \\ (\square\square\square\square\text{X}\square\square\square\square\square\square\square\square)\square\square\square\square \\ \square \\ \square\square\square\square\square\square\square \end{array}$$
[illegible]

☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐☐

☐☐TiCl₄ ☐☐Ti(OC₂H₅)₄ ☐☐Ti(OC₄H₉)₄

☐☐Ti(OC₄H₉)Cl₃ ☐☐☐☐

□0020□

[illegible]
$$\text{Ti}(\text{OC}_2\text{H}_5)_3\text{Cl}_3.$$
$$\text{Ti}(\text{OC}_5\text{H}_5)_2\text{Cl}_2$$

You can list Ti (OC₂ H₅)
₃ Cl, Ti (O-iC₃ H₇)
₃ , Ti (O-nC₄ H₉)
₃ , Ti (O-nC₄ H₉)
₂ Cl₂ , Ti (OC₂ H₅) Br₃ , Ti (OC₂ H₅) (OC₄ H₉)
₂ Cl, Ti (O-nC₄ H₉)
₃ Cl, Ti (O-C₆ H₅)
₃ , Ti (O-iC₄ H₉)
₂ Cl₂ , Ti (OC₅ H₁₁) C₁₃ , Ti (OC₆ H₁₃) C₁₃ , Ti (OC₂ H₅)
₄ , Ti (O-nC₃ H₇)₄ , Ti (O-nC₄ H₉)₄ , Ti (O-iC₄ H₉)₄ , Ti (O-nC₆ H₁₃)₄ , Ti (O-nC₈ H₁₇)₄ , Ti {OCH₂ CH (C₂ H₅) C₄ H₉ }₄ etc.

[0019]

In addition, electron donor which it mentions later in TiX^*_{4} (Here as for X^* halogen is shown.) the molecular compound which reacts can be used.

As embodiment, you can list TiCl_4 * $\text{CH}_3\text{COC}_2\text{H}_5$, TiCl_4 * $\text{CH}_3\text{CO}_2\text{C}_2\text{H}_5$, $\text{C}_2\text{H}_5\text{NO}_2$, TiCl_4 * CH_3COCl , TiCl_4 * $\text{C}_6\text{H}_5\text{COCl}$, TiCl_4 * $\text{C}_6\text{H}_5\text{CO}_2\text{C}_2\text{H}_5$, TiCl_4 * ClCOC_2H_5 , TiCl_4 * $\text{C}_4\text{H}_4\text{O}$ etc.

Even in these titanium compound desirable ones are
 TiCl_4 , $\text{Ti}(\text{OC}_2\text{H}_5)_4$, $\text{Ti}(\text{OC}_4\text{H}_9)_4$, $\text{Ti}(\text{OC}_4\text{H}_9)_3\text{Cl}$ etc.

[0020]

As halogen source, it is normal to be supplied from halogen compound of the above-mentioned magnesium and/or titanium, but it is possible also to supply from the halogenating agent of public knowledge such as halide of aluminum and halide of

the halide , phosphorus of silicon .

halogen which is included in catalyst component fluorine , chlorine , bromine , iodine or is good even with these blend , especially chlorine is desirable.

[0021]

As for solid component which is used for this invention, also use of SiCl_4 , CH_3
 SiCl_3 or other silicon compound, methyl
hydrogen polysiloxane or other polymeric silicon compound,
 $\text{Al}(\text{O}i\text{C}_3\text{H}_7)_3$, AlCl_3 , AlBr_3 , $\text{Al}(\text{OC}_2\text{H}_5)_3$, $\text{Al}(\text{OCH}_3)_2$ Clor other aluminum
compound and $\text{B}(\text{OCH}_3)_3$, $\text{B}(\text{OC}_2\text{H}_5)_3$, $\text{B}(\text{OC}_6\text{H}_5)_3$ or other
borated compound, WCl_6 , MoCl_5
or other other component being possible to other than above-
mentioned essential ingredient, these as forremaining in solid
component as silicon, aluminum and boron or other
component there is not aninconvenience.

Furthermore, when this solid component is produced, using electron donor as the internal donor, it is possible also to produce.

[0022]

esters, ethers, diether of alcohols, phenols, ketones, aldehydes, carboxylic acid and organic acid or inorganic acid and nitrogen-containing electron donor etc like oxygen containing electron donor, ammonia, amine, nitrile, isocyanate like acyl amides, acid anhydride are illustrated as possible electron donor which can be utilized in production of this solid component (internal donor) as.

[0023]

More concrete,

(J2) methanol ,

ethanol ,

propanol ,

pentanol ,

hexanol ,

octanol ,

dodecanol ,

octadecyl alcohol ,

benzyl alcohol ,
phenylethyl alcohol ,
cumyl alcohol ,
alcohols , of isopropyl benzyl alcohol or other carbon number 1 through 18
(jp2) phenol ,
cresol ,
xylenol ,
ethyl phenol ,
propyl phenol ,
cumyl phenol ,
nonylphenol ,
carbon number 6 which is possible to possess naphthol or other alkyl group or phenols , of 25
aldehydes , (jp5) methyl formate , methyl acetate , ethylacetate , vinyl acetate , propyl acetate , octyl acetate , cyclohexyl acetate , cellosolve acetate , ethyl propionate , methyl butyrate , ethyl valerate , ethyl stearate , methyl chloroacetate , ethyl dichloroacetate , methyl methacrylate , ethyl crotonate , ethyl cyclohexane carboxylate , methyl benzoate , ethyl benzoate , propyl benzoate , butyl toluate , octyl benzoate , cyclohexyl benzoate , phenyl benzoate , benzyl benzoate , cellosolve benzoate , methyl toluate , ethyl toluate , amyl toluate , ethyl ethylbenzoate , methyl anisate , ethyl anisate , ethyl ethoxybenzoate , diethyl phthalate , dibutyl phthalate , diheptyl phthalate , ;ga of ketones , (jp4) acetaldehyde , propionaldehyde , octyl aldehyde , benzaldehyde , tolualdehyde , naphthaldehyde or other carbon number 2 to 15 of (jp3) acetone , methylethyl ketone , methyl isobutyl ketone , acetophenone , benzophenone or other carbon number 3 to 15 -butyrolactone , the;al -valerolactone , coumarin , phthalide , ethylene carbonate or other carbon number 2 to 20 organic acid esters , acid halide , (jp8) methyl ether , ethyl ether , isopropyl ether , butyl ether , amyl ether , tetrahydrofuran , anisol , diphenylether or other carbon number 2 to 20 ethers , 2,2- diisopropyl -1,3- dimethoxy propane , 2- isopropyl -2- isopentyl -1,3- dimethoxy propane , 2- phenyl -2- s-butyl -1,3- dimethoxy propane or other diether of inorganic acid esters , (jp7) acetyl chloride , benzoyl chloride , toluoyl chloride , anisoyl chloride , phthaloyl chloride , isophthaloyl chloride or other carbon number 2 to 15 like (jp6) ethyl silicate , butyl silicate , phenyl triethoxysilane or other silicate ester and (jp9) acetic acid amide , benzoic acid amide , toluamide or other acyl amides , (jp10) methylamine , ethylamine , diethylamine , tributyl amine , piperidine , tri benzylamine , aniline , pyridine , picoline , tetramethyl ethylenediamine or other amines , (jp11) acetonitrile , benzonitrile , tolunitrile or other nitriles , etc

can be listed.

2 kinds or more you can use these electron donor.

As for being desirable among these with organic acid ester and organic acid halide , and diether , as for especially being desirable it is a phthalic acid ester , cellosolve acetate , phthalic acid halide and diether .

[0024]

As for amount used of above-mentioned each component, it can recognize effect of this invention, if it can with those of option, but generally, inside of following range is desirable.

As for amount used of titanium compound, inside of range of $1 \times 10^{-4} \sim 1000$ is good with mole ratio vis-a-vis amount used of magnesium compound which is used, it is inside range of preferably 0.01~10.

When compound for that is used as halogen source, amount used titanium compound and/or magnesium compound includes halogen, it is inside range of $1 \times 10^{-2} \sim 1000$, preferably $0.1 \sim 100$, with mole ratio vis-a-vis amount used of magnesium which is used it does not include of regardless.

[0025]

amount used of silicon , aluminum and borated compound is inside range of $1 \times 10^{-3} \sim 100$, preferably $0.01 \sim 1$, with mole ratio vis-a-vis amount used of above-mentioned magnesium compound .

amount used of electron donating compound is inside range of $1 \times 10^{-3} \sim 10$, preferably 0.01~5, with mole ratio vis-a-vis amount used of above-mentioned magnesium compound .

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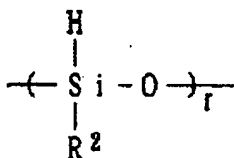
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[0026]

solid component in order to produce component (A) above-mentioned titanium source, magnesium source and halogen source, furthermore is produced making use of electron donor or other other component, likebelow for example by production method in accordance with necessary.

(J2) magnesium halide and according to need electron donor and titanium-containing compound method. which contacts

method. which treats (jp2) alumina or magnesia with halogenated phosphorus compound, the magnesium halide, electron donor, titanium halogen containing compound contacts that

(jp3) magnesium halide and titanium tetraalkoxide and specific polymeric silicon compound contacting, in the solid component which is acquired, halogen compound, and according to need electron donor of titanium halogen compound and/or silicon the method. which contacts

As this polymeric silicon compound, those which are shown with formula below are suitable.

[0027]

[Chemical Formula 1]

[0028]

(Here, as for R² as for hydrocarbon residue, r of carbon number 1~10extent, kind of degree of polymerization where viscosity of this polymeric silicon compound becomes 1 - 100 centistokes extent is shown.)

Among these, methyl hydrogen polysiloxane, 1,3,5,7-tetramethyl cyclotetrasiloxane, 1,3,5,7,9-pentamethyl cyclopentasiloxane, ethyl hydrogen polysiloxane, phenyl hydrogen polysiloxane, cyclohexyl hydrogen polysiloxane etc is desirable.

[0029]

Melting (jp4) magnesium compound with titanium tetraalkoxide and electron donor, method. which the titanium compound contacts solid component which it precipitated

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with halogenating agent or the titanium halogen compound

(jp5) Grignard reagent or other organomagnesium compound
halogenating agent , reductant etc and after action, in this
according to need electron donor and the titanium compound
method . which contacts

In (jp6) alkoxy magnesium compound halogenating agent
and/or titanium compound method . which contacts under
existing or absence of electron donor

[0030]

As manufacturing condition of component (A) of this
invention , inside ofbelow-mentioned range is desirable.

contact temperature - is 50 - 200 deg Cextent , preferably
0~100 deg Cextent , .

As contact method , mechanical method with such as rotating
ball mill , vibrating mill , jet mill , medium-stirring mill .
Under existing of inactivity diluent , you can list method etc
which contactswith churning.

You can list hydrocarbon and halohydrocarbon , polysiloxane
etc of aliphatic or aromatic as inactivity diluent which this
time is used.

[0031]

component (A) which is used with this invention can also use
as those whichpass prepolymerization step which consists of
fact that contacting, youpolymerize vinyl group-containing
compound , for example olefins , diene compound , styrene
etc.

As embodiment of olefins which is used occasion where
prepolymerization is done, those of for example carbon
number 2~20extent . There is a ethylene , propylene , 1-
butene , 3- methyl butene -1,1- pentene , 1- hexene , 4-
methylpentene -1,1- octene , 1- decene , 1- undecene , 1-
eicosene etc concretely, there are 1 and 3 -butadiene ,
isoprene , 1,4- hexadiene , 1,5-hexadiene , 1,3- pentadiene ,
1,4- pentadiene , 2,4- pentadiene , 2,6-octadiene , cis-2,trans -
4- hexadiene , trans -2, trans -4- hexadiene , 1,3- heptadiene ,
1,4- heptadiene , 1,5-heptadiene , 1,6-heptadiene , 2,4-
heptadiene , dicyclopentadiene , 1,3- cyclohexadiene , 1,4-
cyclohexadiene , cyclopentadiene , 1,3- cyclo heptadiene , 4-
methyl -1,4- hexadiene , 5-methyl -1,4- hexadiene , 1,9-
decadiene , 1,13- tetradecadiene , p- divinyl benzene , m-
divinyl benzene , o-divinyl benzene , dicyclopentadiene etc as
the embodiment of diene compound .

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$\text{AlR}^{3-3-n} \text{X}^{1-n} \text{I} \text{I}$

In addition, you can list styrene , al -methylstyrene , allyl benzene , chlorostyrene etc as embodiment of the styrene .

[0032]

As for reaction condition of titanium component and above-mentioned vinyl group-containing compound , it can recognize effect of this invention , if it can with those of option ,but inside of following range is desirable generally.

amount of prepolymerization of vinyl group-containing compound per titanium solid component 1gram 0.001 - 1000 gram , preferably 1~100gram , furthermore is inside range of preferably 5~50gram .

reaction temperature at time of prepolymerization - is 150 - 85 deg C, preferably 0~50 deg C.

Low polymerization temperature is desirable and, "This polymerization ", namely the al -olefin in comparison with polymerization temperature when polymerizing.

Reaction does generally under agitating, it is desirable , that time n- hexane , n-heptane or other inert solvent to be possible also, it is possible also, in addition with types of vinyl compound to do that itself of above-mentioned vinyl compound to exist, as media it is possible also to execute with the gas state .

In addition 2 kinds or more also to jointly use this vinyl group-containing compound it is possible.

In addition, as for melting point of polymer which prepolymerization is done, it is desirable from reason of activity improvement of component (A) to be 200 deg C or greater .

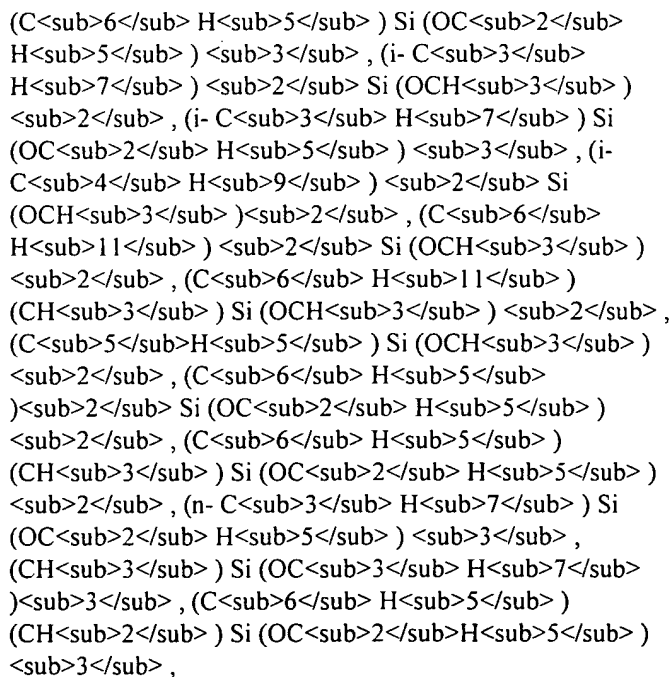
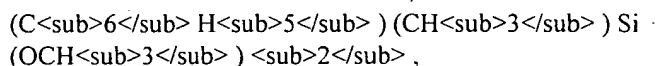
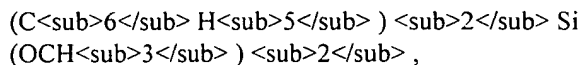
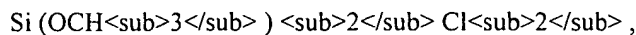
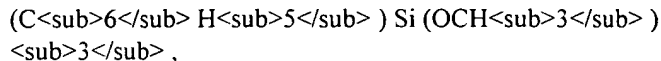
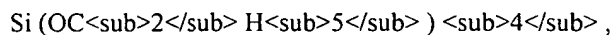
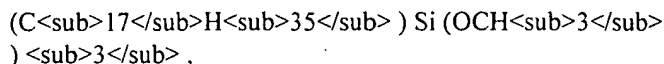
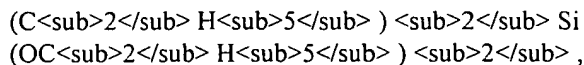
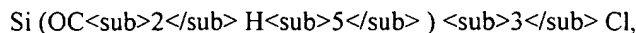
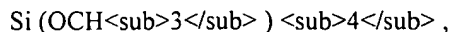
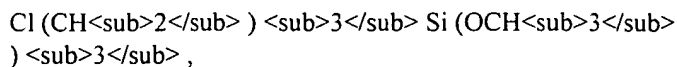
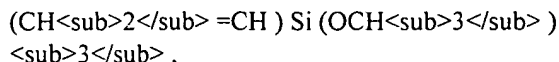
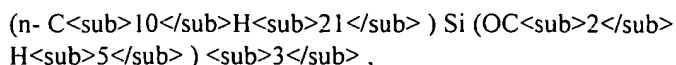
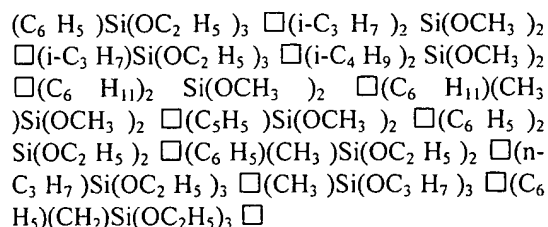
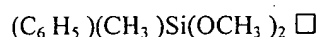
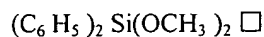
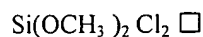
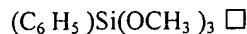
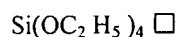
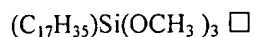
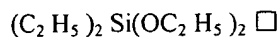
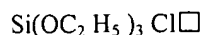
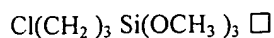
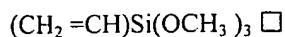
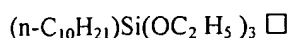
[0033]

As for component (B) of <component (B)> this invention , blend , of organo-aluminum compound , or organo-aluminum compound which is displayed with below-mentioned General Formula [II] and is displayed with below-mentioned General Formula [III] organo-aluminum compound which are displayed with below-mentioned General Formula [I]

$\text{AlR}^{3-3-n} \text{X}^{1-n} \text{I} \text{I}$

$\square\square\square\square\text{I}\square\square\square\square\square\square\square\square\square\square\square\square$
 $\square\square\square\square\square\square\square\square\square\square\text{Al}(\text{CH}_3)_3 \quad \square\text{Al}(\text{C}_6\text{H}_5)_3 \quad \square\text{Al}(\text{i-C}_4\text{H}_9)_3 \quad \square\text{Al}(\text{C}_4\text{H}_9)_3 \quad \square\text{Al}(\text{n-C}_6\text{H}_{13})_3 \quad \square\text{Al}(\text{n-C}_8\text{H}_{17})_3 \quad \square\text{Al}(\text{C}_{10}\text{H}_{21})_3 \quad \square(\text{CH}_3)_2\text{AlCl} \quad \square(\text{C}_2\text{H}_5)_2\text{AlCl} \quad \square(\text{C}_2\text{H}_5)_3\text{Al}_2\text{Cl}_3 \quad \square(\text{C}_2\text{H}_5)\text{AlCl}_2 \quad \square(\text{n-C}_3\text{H}_7)_2\text{AlCl} \quad \square(\text{i-C}_3\text{H}_7)_2\text{AlCl} \quad \square(\text{n-C}_4\text{H}_9)_2\text{AlCl} \quad \square(\text{i-C}_4\text{H}_9)\text{AlCl}_2 \quad \square(\text{n-C}_6\text{H}_{13})_2\text{AlCl} \quad \square(\text{n-C}_8\text{H}_{17})_2\text{AlCl} \quad \square(\text{n-C}_{10}\text{H}_{21})_2\text{AlCl} \quad \square\square\square(\text{C}_2\text{H}_5)_2\text{AlBr} \quad \square\square\square\square\square$

There is a Al (CH₃)₃ , Al (C₂H₅)₃ , Al (iC₄H₉)₃ , Al (C₄H₉)₃ , Al (n-C₆H₁₃)₃ , Al (n-C₈H₁₇)₃ , Al (C₁₀H₂₁)₃ , (CH₃)₂ Al Cl, (C₂H₅)₂ Al Cl, (C₂H₅)₃ Al , Cl₃ , (C₂H₅)₃ Al , Cl₂ , (n- C₃H₇)₂ Al Cl, (i- C₃H₇)₂ Al Cl, (n- C₄H₉)₂ Al Cl, (i- C₄H₉)₂ Al Cl₂ , (n- C₆H₁₃)₂ Al Cl, (n- C₈H₁₇)₂ Al Cl, (n- C₁₀H₂₁)₂ Al Cl,

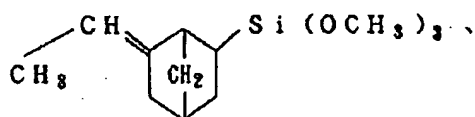
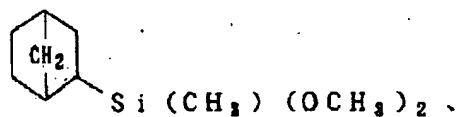
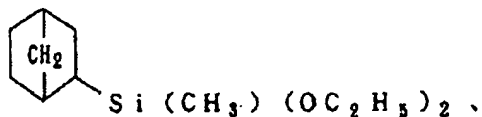
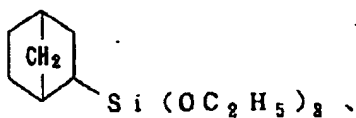


[0042]

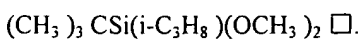
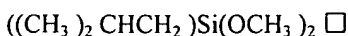
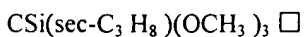
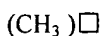
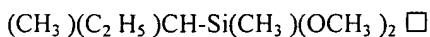
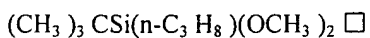
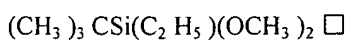
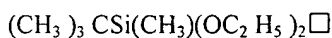
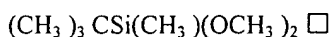
[Chemical Formula 2]

□0042□

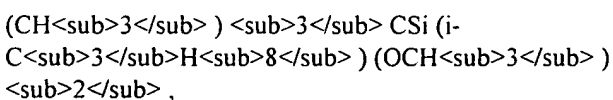
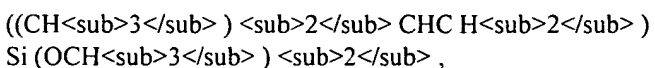
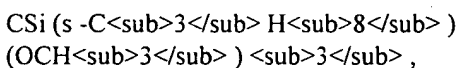
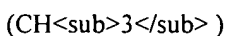
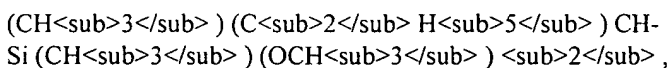
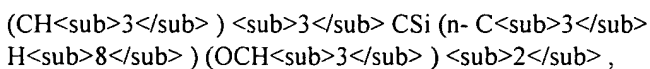
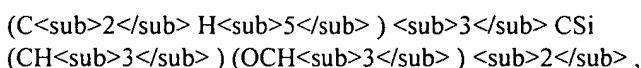
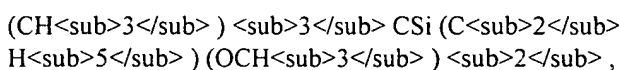
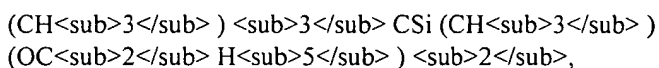
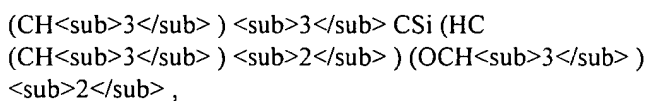
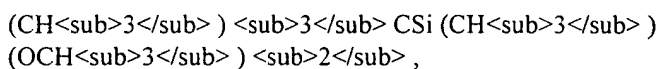
□□2□



□0043□



[0043]



[illegible][illegible]

□0044□

[illegible]

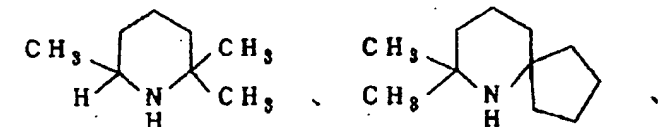
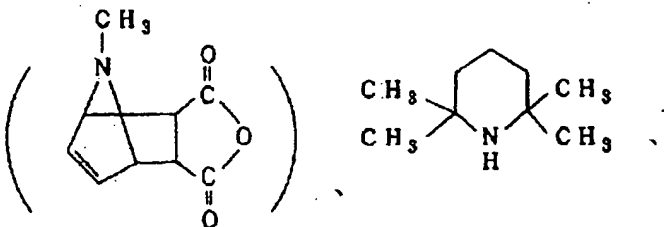
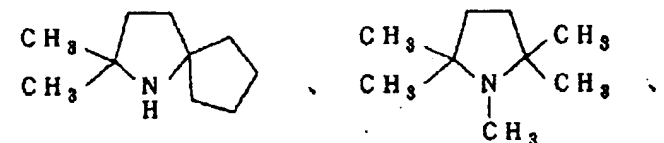
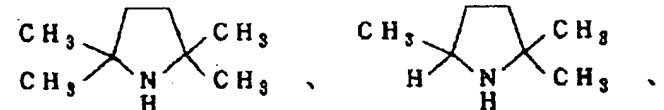
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 (□□□□□□□□)□□□2,2,6,6-
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0045

□□3□



□0046□

0040

etc.

Fact that it is desirable among these, carbon of the α position being secondary or tertiary, carbon of carbon number 3~20, particularly preferably α position being tertiary, is silicon compound which possesses branched chain hydrocarbon residue of carbon number 4~12.

Here, "branched chain " with, branched aliphatic and cycloaliphatic are included.

[0044]

As amines , generally known "hindered amine " is suitable.

Especially N- substitution (As for substituent lower alkyl group or lower alkyl aluminum group) or unsubstituted pyrrolidine or piperidine 2, 2, 5 and 5 - (When of pyrrolidine) or 2, 2, 6 and 6 -tetra lower alkyl substituted compound are representative .

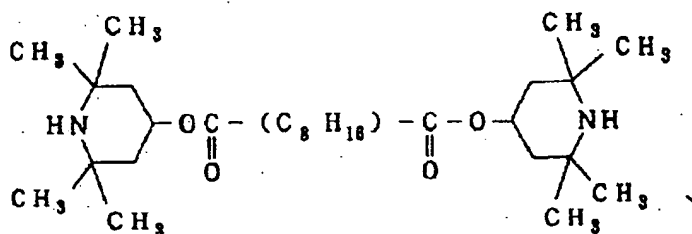
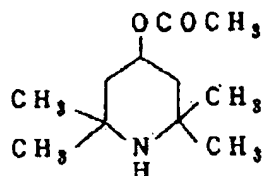
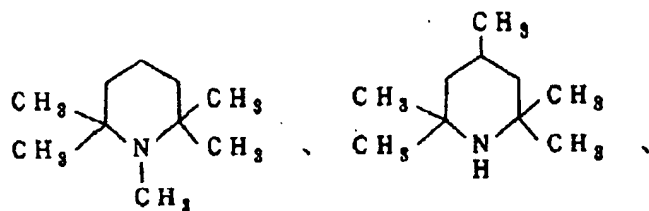
Below-mentioned ones can be illustrated as that kind of amines .

[0045]

[Chemical Formula 3]

[0046]

[Chemical Formula 4]



□0047□

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 □□□□□2□□□□□□□□□□□□□6
 □□□□□□□□□□□1□□□□□□□□
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□0048□

□□5□

[0047]

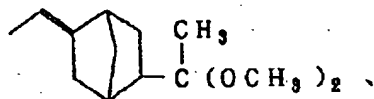
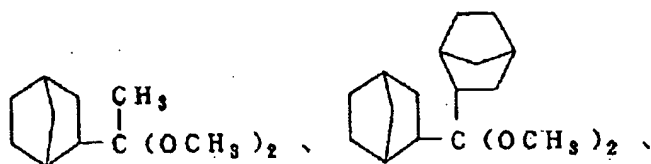
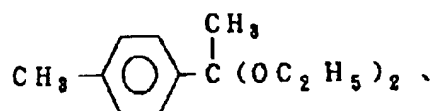
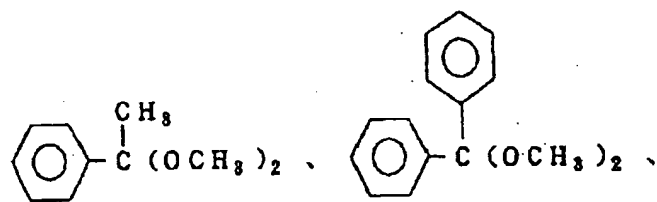
As ethers, those which it can call hindered ether are suitable.

Especially, at least two it possesses lower alkoxy group on carbon atom of the one, this said carbon atom furthermore those which at least one it possesses 6-member ring substituent is representative.

Below-mentioned ones can be illustrated as that kind of ethers

[0048]

[Chemical Formula 5]

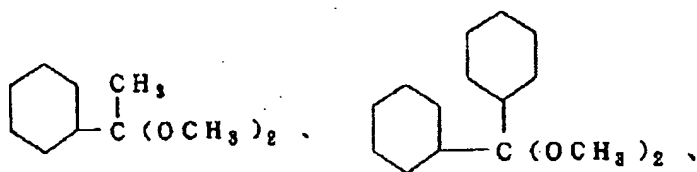
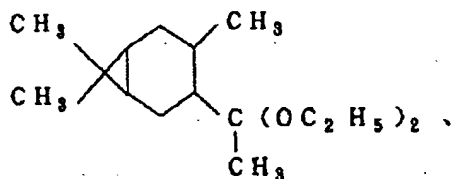
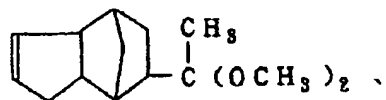
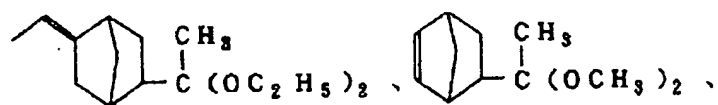


□0049□

[0049]

□□6□

[Chemical Formula 6]

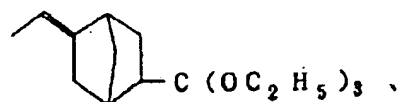
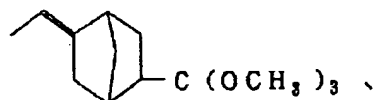
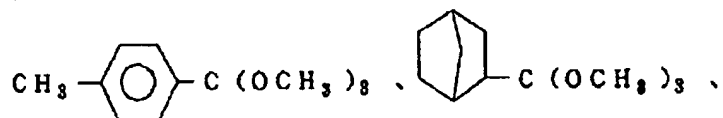
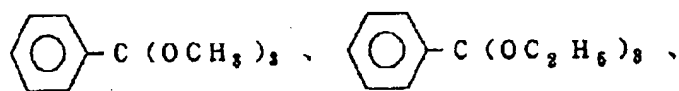


□0050□

[0050]

□□7□

[Chemical Formula 7]

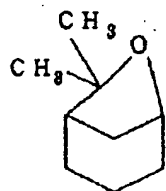
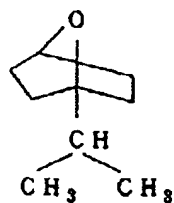
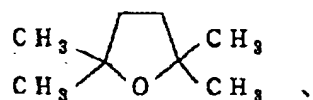
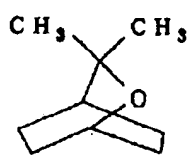
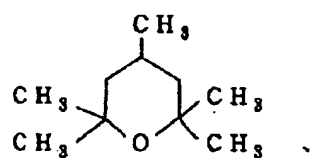
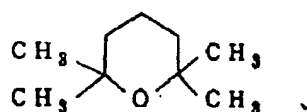
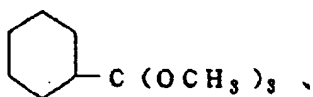
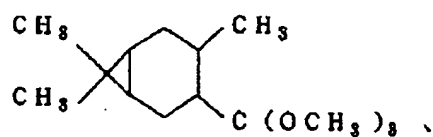
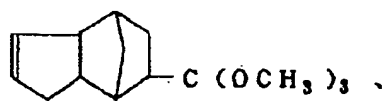


□0051□

[0051]

□□8□

[Chemical Formula 8]



□0052□

□□□□□□□□(C)□□□□□□□□□□
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□□□□□1,3-
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 2-□□□□□□-1,3-□□□□□□□□□□□□2-
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 1,3-□□□□□□□□□□2,2-
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 1,3-□□□□□□□□□□2,2-

[0052]

With this invention, it can also use diether compound component (C) as.

If it is a diether compound, there is not restriction. Desirable ones are diether which possesses branched aliphatic hydrocarbon and/or cycloaliphatic hydrocarbon group even among them.

Among these, it can use 1 and 3 -diether desirably, especially, 2 and 2-diisobutyl -1,3- dimethoxy propane, 2- isopropyl -2- isopentyl -1,3- dimethoxy propane, 2,2- dicyclohexyl -1,3- dimethoxy propane, 2,2- bis (cyclohexyl methyl) - 1 and 3 - dimethoxy propane, 2,2- diisopropyl -1,3- dimethoxy propane, 2,2- dicyclo pentyl -1,3- dimethoxy propane, 2- cyclopentyl -2- isopropyl -1,3- dimethoxy propane, 2- cyclopentyl -2- s-butyl -1,3- dimethoxy propane, 2- cyclohexyl -2- isopropyl -1,3- dimethoxy propane, 2-

□□□ 1

Working Example 1

□□□(A)□□□□□□□□□□□□□□
 □□□□□□□□□□□□□□□□□□100
 □□□□□□□□□□□□□□□□Mg(OEt)₂
 □10□□□□□□□□□□□□□□□□

□□□□TiCl₄ 20□□□□□□□□□□□90
 deg C□□□□□2-□□□□□□-2-
 □□□□□□-1,3-
 □□□□□□□□1.9□□□□□□□□□□
 □□110 deg C□□□□□3□□□□□□□□

□□□□□□□□□□□□□□□□

□□□TiCl₄
 20□□□□□□□□□□□□□□100□□□□
 □□□□□□110 deg
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 C□□□□□□

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 □□□□□□180 deg C□100kg/cm²
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In flask which nitrogen substitution is done, dehydration and 10 gram and and suspension state it introduced toluene 100ml which deoxygenation is done in {Production of component (A)} satisfactory, next Mg (OEt)₂ introduced made.

Next, it introduced TiCl₄ 20ml, temperature rise did in 90 deg C and introduced 2-isopropyl-2-isopentyl-1,3-dimethoxy propane 1.9ml, temperature rise did in 110 deg C and 3 hours reacted.

After reaction termination, you washed with toluene.

Next, it introduced TiCl₄ 20ml and toluene 100ml, 2 hours reacted with 110 deg C.

After reaction termination, with n-heptane washing in satisfactory, solid component in order to produce component (A) (A-1) with it did.

This titanium content was 2.5 weight %.

[0056]

Next, in autoclave which nitrogen substitution is done, n-heptane which was refined in same way as description above 200 ml was introduced in satisfactory, solid component which is synthesized at description above 5 gram was introduced, triethyl aluminum 1.5 gram was introduced next, 4-methylpentene-1 the prepolymerization was executed with 20 deg C.

After prepolymerization ending, with n-heptane washing in satisfactory, component (A) with it did.

Removing portion, when you inspected amount of prepolymerization they were 10.6 gram poly 4-methylpentene-1/gram solid component (A-1).

Furthermore melting point of poly 4-methylpentene-1 was 231 deg C.

[0057]

In stainless steel autoclave of internal volume 1.0 liter which possesses {Polymerization of propylene} churning and temperature control equipment in satisfactory dehydration and propylene which deoxygenation is done triethyl aluminum 8.9 milligram was introduced with 25 deg C 100 ml, component (B) as.

While introducing propylene into autoclave, temperature rise, increased pressure it did and it designated reaction system as supercritical state to 180 deg C, 100kg/cm².

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This titanium content was 2.6 weight %.

[0060]

Next, in autoclave which nitrogen substitution is done, n-heptane which was refined in same way as description above 200 ml was introduced in satisfactory, solid component which is synthesized at description above 5 gram was introduced, triethyl aluminum 1.5 gram was introduced next, 3-methyl butene -1 the prepolymerization was executed with 10 deg C.

After prepolymerization ending, with n-heptane washing in satisfactory, component (A) with it did.

Removing portion, when you inspected amount of prepolymerization they were 12.7 gram poly 3-methyl butene -1/gram solid component (A-1).

Furthermore melting point of poly 3-methyl butene -1 was 302 deg C.

[0061]

propylene was introduced into autoclave in same way as {Polymerization of propylene} Working Example -1.

Next (CH₃)₃CSi(n-C₃H₇)(OCH₃)₂ 1.0 milligram, hydrogen 5 ml was introduced next triethyl aluminum 4.7 milligram, component (C) as component (B) as.

While introducing propylene into autoclave, it did temperature rise, increased pressure and it designated reaction system as supercritical state to 200 deg C, 150 kg/cm².

Next 3 milligram it introduced component (A) which is produced at description above with component (A-1) standard and started the polymerization and 45 min polymerized and stopped reaction in the same way as Working Example -1.

polymer of 127 gram was acquired, with MFR = 68.2 g/10 min, it was a I.I = 97.1 wt% with boiling n-heptane extraction.

In addition, Q was 7.1.

[0062]

Working Example 3~6

Using component (A) which is produced with Working Example -2, component which it shows in Table 1 (B) and using component (C), other than making the polymerization condition which it shows in Table 1, you polymerized with condition which is completely similar to Working Example 2.

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Result is shown in Table 1 .

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[0063]

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[Table 1]

表 1

実施例	成分 (B) (使用量)	成分 (C) (使用量)	単量体成分		重合時		重合結果			
			臨界温度	臨界圧力	温度 (°C)	圧力 (kg/cm ²)	収率-収量 (g)	MER (g/10分)	I. I (wt%)	Q値
実施例 1	トリメチルシリル (8.9mg)	—	70℃ 91.4℃	45.4kg/cm ²	180	100	104	51.8	95.3	6.2
実施例 2	トリメチルシリル (4.7mg)	(CH ₃) ₃ CSi(H-C ₈ H ₇ OCH ₃) ₂ (1mg)			200	150	127	68.2	97.1	7.1
実施例 3	トリメチルシリル (12mg)	$ \begin{array}{c} \text{CH}_3 \text{ CH}_3 \\ \quad \\ \text{H}_3\text{C} - \text{C} - \text{Si} - \text{OCH}_3 \\ \quad \quad \\ \text{CH}_3 \text{ CH}_3 \text{ nC}_8\text{H}_7 \end{array} $ (1.3mg)			190	135	125	51.6	96.8	7.3
実施例 4	トリメチルシリル (11.5mg)	$ \begin{array}{c} \text{t-C}_4\text{H}_9 \\ \diagup \quad \diagdown \\ \text{Si} - \text{OCH}_3 \\ \diagdown \quad \diagup \\ \text{n-C}_4\text{H}_9 \end{array} $ (1.1mg)			185	140	129	34.7	97.3	7.6
実施例 5	トリメチルシリル (18.5mg)	(C ₂ H ₅) ₂ Si(OCH ₃) ₂ (1.9mg)			200	140	115	88.3	95.4	7.0
実施例 6	トリメチルシリル (27.3mg)	$ \begin{array}{c} \text{t-C}_4\text{H}_9 \\ \diagup \quad \diagdown \\ \text{Si} - \text{OC}_2\text{H}_5 \\ \diagdown \quad \diagup \\ \text{CH}_3 \quad \text{OC}_2\text{H}_5 \end{array} $ (2.5mg)			210	170	111	117.2	95.2	8.8

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[Brief Explanation of the Drawing(s)]

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[Figure 1]

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It is a flowchart in order to help understanding of this invention .

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Drawings

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[Figure 1]

